A Top-Down Emissions Inventory Evaluation for the Upper Midwest

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INTRODUCTION

This study, funded by the National Renewable Energy Laboratory (NREL), focused on a top-down evaluation of the 2005 emissions inventory that the Lake Michigan Air Directors Consortium (LADCO) is using to conduct regional air quality modeling in the upper Midwest. The on-road mobile source component of the inventory was a special focus of this study.

Several techniques can be used to evaluate the accuracy of any emissions inventory that is intended for use in air quality modeling: "common sense" review of the data; bottom-up evaluations that start with emissions activity data to estimate corresponding emissions; and top-down evaluations that compare emissions estimates to ambient air quality data. As a top-down emissions inventory evaluation, this work focused on comparing the LADCO emissions inventory to ambient monitoring data collected at four urban areas in the region of interest: Chicago, Illinois; Milwaukee, Wisconsin; Gary, Indiana; and Detroit, Michigan.

The goals of the study were to (1) identify areas of agreement and differences between the ambient data and emissions inventory; (2) identify areas of the emissions inventory that may need improvement; and (3) demonstrate the usefulness of top-down emissions inventory evaluation techniques.

Locations of monitoring sites included in the top-down emissions inventory evaluation.

TECHNICAL APPROACH

Top-down emissions evaluations can be confounded by the fact that ambient concentrations are influenced not only by fresh, local emissions, but also by transported pollution and chemical reactions occurring after pollutants are emitted. To minimize the influence of these effects, we used data from early morning periods (0600-0900) when emission rates are high and reaction rates are low.

It should also be noted that, due to the inherent uncertainties associated with top-down evaluations, ambient- and emissions inventory-derived pollutant ratios within 25-50% of each other are considered to be in good agreement.

Data Acquisition and Processing

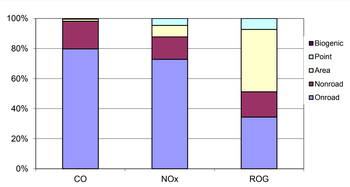
As part of this study, STI

- Acquired air quality and meteorological data from the U.S. EPA's Air Quality System for the monitoring sites. Hourly data were acquired for the years 2004-2006 for summer (June-August) and winter (December-February) months.
- Acquired LADCO's 2005 Base M emissions inventory data and supporting files (e.g., speciation profiles) for area, point, nonroad mobile, on-road mobile, and biogenic sources.
- Speciated the 2005 emissions data and matched individual hydrocarbon species with those measured in the ambient data.
- Converted emissions data from mass to molar units.

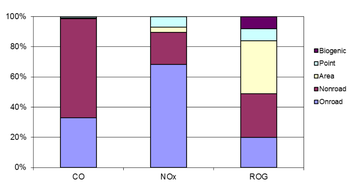
Site Emissions Characterization

To characterize the source mix around each monitoring site, emissions data were summarized for each site's grid analysis zone.

- On-road mobile sources accounted for 61-80% of winter CO emissions and 57-73% of winter and summer NO_x emissions at all sites except for Gary.
- Point sources accounted for the majority of CO and NO_x emissions for both seasons at the Gary site.
- Area sources accounted for 35-57% of the winter and summer volatile organic compound (VOC) emissions at each site, while on-road mobile sources accounted for 15-39% of the winter and summer VOC emissions at each site.



January weekday emissions by source sector for the area around the Chicago-Mannheim site.



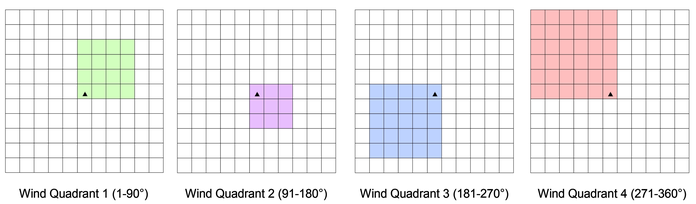
July weekday emissions by source sector for the area around the Chicago-Mannheim site.

Criteria Pollutant Ratios

Total non-methane organic compound (TNMOC)/NO_x and CO/NO_x ratios from the ambient and emissions inventory data were computed and compared by

- Day of week
- Month
- Season (winter/summer)
- Wind quadrant (defined below)

For the gridded (4 km x 4 km) emissions inventory, grid analysis zones around each site were identified based on predominant winds during the early morning hours (0600-0900). In addition, groups of cells associated with individual wind quadrants were identified at each site; the spatial extent of each wind quadrant varied according to the observed winds for that quadrant and site.



Example of a full extent grid analysis zone, showing the spatial configuration of grid cells for which ambient- and emissions inventory-derived ratio comparisons were calculated. The hollow grid represents the entire grid analysis zone and the colored regions represent the wind quadrant analysis zones.

Hydrocarbon Composition

The chemical composition of hydrocarbons reported in the emissions inventory was compared to the chemical composition of the ambient air at individual monitoring sites. These "fingerprint" analyses were used to evaluate the accuracy of the speciation of the emissions inventory.

In addition, the relative reactivity of the organic species in the emissions inventory and ambient data were computed and compared. Weighted reactivity values for the emissions inventory and ambient data were calculated as follows:

$$R = \sum (MIR)_i w_i$$

where:

R = weighted reactivity
$(MIR)_i$ = maximum incremental reactivity for species *i*
w_i = weight fraction of species *i* in the emissions inventory or ambient data

RESULTS

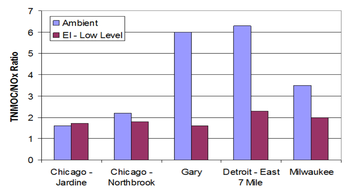
TNMOC/NO_x Ratios

Summer TNMOC/NO_x ratio comparisons were made for five sites: Chicago-Jardine, Chicago-Northbrook, Gary, Detroit-East 7 Mile, and Milwaukee. These comparisons show good agreement (±20%) between ambient- and emissions inventory-derived ratios at the Chicago sites; however, at the remaining sites, ambient-derived ratios were higher than emissions inventory-derived ratios by a factor of 1.8 or more.

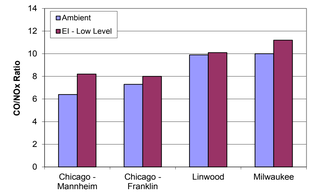
In addition, agreement between ambient- and emissions inventory-derived TNMOC/NO_x ratios at the Chicago sites is poorer on weekend days than on weekdays. This is particularly true for Sundays at the Northbrook site, where the ambient-derived TNMOC/NO_x ratio is two times higher than the emissions inventory-derived ratio.

CO/NO_x Ratios

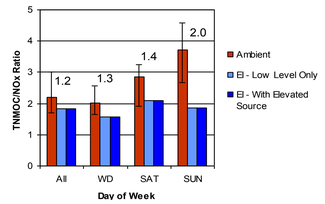
Summer and winter CO/NO_x ratio comparisons were made for four sites: Chicago-Mannheim, Chicago-Franklin, Detroit-Linwood, and Milwaukee. These comparisons showed that ambient- and emissions inventory-derived ratios had close agreement at all sites (±20% for winter comparisons). However, at the Chicago sites, the agreement is poorer on Sundays than on Saturdays and weekdays.



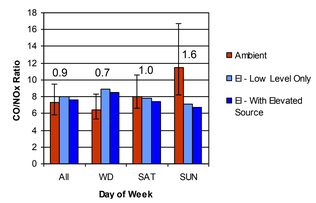
Summer ambient- and emissions inventory-derived TNMOC/NO_x ratios by site.



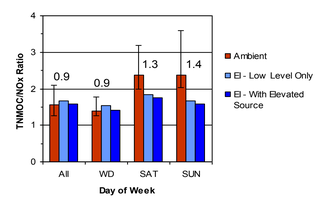
Winter ambient- and emissions inventory-derived CO/NO_x ratios by site.



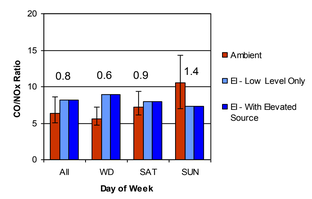
Summer TNMOC/NO_x ratios by day of week for the Chicago-Northbrook site.



Winter CO/NO_x ratios by day of week for the Chicago-Franklin site.



Summer TNMOC/NO_x ratios by day of week for the Chicago-Jardine site.



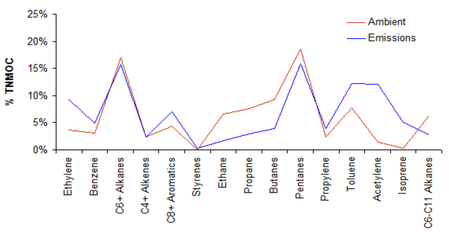
Winter CO/NO_x ratios by day of week for the Chicago-Mannheim site.

Hydrocarbon Compositions

In general, our comparisons of the ambient- and emissions inventory-derived relative hydrocarbon compositions showed that

- There was good agreement for some species (e.g., C4+ alkanes, C8+ aromatics, styrenes, and propylene)
- The contribution of some species is overestimated in the inventory (e.g., acetylene, ethylene, toluene, and isoprene)
- The contribution of some species is underestimated in the inventory (e.g., ethane, propane, and C6-C11 alkanes)

To investigate the potential impact of these speciation issues on ozone formation, the weighted reactivity of the mix of hydrocarbon species in the ambient and emissions inventory data were calculated and compared. Across all sites, the weighted reactivity values for the summer emissions inventory were 16% to 80% higher than the weighted reactivity values for the ambient data.



Comparison of 0600–0900 ambient- and emissions inventory-derived TNMOC compositions for the Chicago-Jardine site.

CONCLUSIONS

The top-down evaluation for the 2005 emissions inventory indicates that, in general, on-road mobile sources are represented accurately in the emissions data. This conclusion is based on the fact that agreement between ambient- and emissions inventory-derived pollutant ratios was closest (±20%) for wintertime CO/NO_x ratios, and on-road mobile sources accounted for 57% to 80% of wintertime CO and NO_x emissions at all sites for which ratios were calculated.

However, comparisons with ambient data indicate that the emissions inventory for weekends (especially Sundays) may not be representative of actual activity patterns, perhaps due to decreases in heavy-duty truck emissions from weekdays to weekends, as well as inaccurate temporal characterizations of other source categories.

Other key findings include:

- The speciation of the VOC emissions inventory at all sites does not compare well with the hydrocarbon composition of the ambient data.
- The resulting VOC emissions inventory is more reactive (i.e., prone to contribute to ozone formation) at all sites than the corresponding ambient data.

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